



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(21) International Application Number:</b> PCT/GB99/03619  <b>(22) International Filing Date:</b> 2 November 1999 (02.11.99)  <b>(30) Priority Data:</b> 9823761.3                      2 November 1998 (02.11.98)                      GB  <b>(71) Applicant (for all designated States except US):</b> SOUTH BANK UNIVERSITY ENTERPRISES LTD. [GB/GB]; 103 Borough Road, London SE1 0AA (GB).  <b>(72) Inventor; and</b> <b>(75) Inventor/Applicant (for US only):</b> KATHIRGAMANATHAN, Poopathy [GB/GB]; 14 Sandhurst Avenue, North Harrow, Middlesex HA2 7AP (GB).  <b>(74) Agent:</b> COHEN, Alan, Nicol; 2 Grove Place, Tatsfield, Westerham, Kent TN16 2BB (GB).		<b>(81) Designated States:</b> AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
<b>(54) Title:</b> ELECTROLUMISCENT MATERIALS  <b>(57) Abstract</b>  A novel electroluminescent compound which emits blue or blueish purple light is a complex of a lanthanide or actinide and an organic complex e.g. mono(bathophenanthroline) thoriumIV chloride or mono(tripyridyl)yttrium chloride.		

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## ELECTROLUMINESCENT MATERIALS

5 The present invention relates to electroluminescent materials and to devices incorporating them.

10 Materials which emit light when an electric current is passed through them are well known and used in a wide range of display applications. Liquid crystal devices and devices which are based on inorganic semiconductor systems are widely used, however these suffer from the disadvantages of high energy consumption, high cost of manufacture, low quantum- efficiency and the inability to make flat panel displays. reflectance problems, i.e. low visibility in bright conditions and a narrow viewing angle e.g. +/- 45°.

15 Organic polymers have been proposed as useful in electroluminescent devices, but it is not possible to obtain pure colours, they are expensive to make and have a relatively low efficiency.

20 Another compound which has been proposed is aluminium quinolate, but this requires dopants to be used to obtain a range of colours and has a relatively low efficiency.

25 In an article in Chemistry letters pp 657-660, 1990 Kido et al disclosed that a terbium (III) acetyl acetonate complex was green electroluminescent and in an article in Applied Physics letters 65 (17) 24 October 1994 Kido et al disclosed that a europium (III) triphenylene diamine complexes was red electroluminescent but these were unstable in atmospheric conditions and difficult to produce as films.

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The complexes disclosed in these articles had a relatively low photoluminescent efficiency and were only able to produce green or red light and other colours could not be produced.

- 5 We have now discovered photoluminescent and electroluminescent compounds and materials which emit blue and purplish blue light.

10 According to the invention there is provided an photoluminescent compound which comprises an organic complex of a transition metal, lanthanide or an actinide and an organic ligand which photoluminescent compound emits light in the blue or purplish blue spectrum.

15 It has surprisingly been found that it is possible by choice of the metal and the organic ligand to form a complex which, when an electric current is applied across it will emit blue or purplish blue light.

20 The invention also provides an electroluminescent compound which comprises an organic complex of a transition metal, a lanthanide or an actinide and an organic ligand which electroluminescent compound emits light in the blue or purplish blue spectrum when an electric current is passed through it.

25 The colour of light is subjective and colours can be defined by co-ordinates on a two dimensional chart in which colours are areas on the chart and in the present invention the blue and purplish blue spectrum is defined as the area bounded by the co-ordinates in the colour chart CIE I931 a copy of which is shown in Fig. 1. The complexes of the invention enable light within the co-ordinates (0, 0) (0, 0.3) (0.3, 0) to be produced.

30 Light in the blue region of the spectrum is difficult to produce and hitherto it has not been possible to produce blue light by means of electroluminescence.

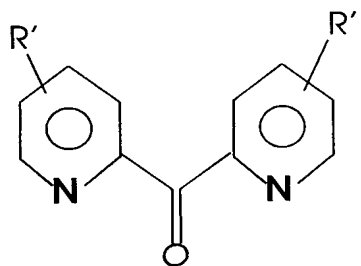
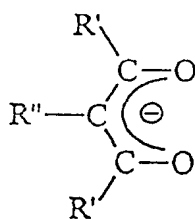
- 3 -

The preferred metals are thorium (IV), yttrium (III), gadolinium (III), europium (II), terbium(IV), cerium(IV) and cerium (III). A mixture of metals can be used to form mixed chelates.

5

The preferred ligands are

10



or

15 where R' is the same or different at different parts of the molecule and each R'' and R' is a substituted or unsubstituted aromatic or heterocyclic ring structure or a hydrocarbyl or a fluorocarbon or R'' is fluorine or hydrogen or R'' is copolymerised with a monomer e.g. or R' is t-butyl and R'' hydrogen.

20 Preferably each of R', R'', and R' is an alkyl group preferably a -C(CH<sub>3</sub>) group,

Preferred complexes are TMHD (Tris(2,2,6,6-tetramethyl-3,5-heptanedionato). α', α'', α''' tripyridyl, bathophen (4,7-diphenyl-1,1-phenanthroline), crown ethers and cryptans.

25

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Particularly preferred complexes are the thorium bathophen, yttrium tripyridyl and TMHD, and europium TMHD complexes.

5 A novel complex with strong photoluminescent and electroluminescent properties is  $\text{Eu(II)(TMHD)}_2$  complex which is stable in air. It would have been expected that such a Eu(II) complex would have been unstable in the presence of oxygen and it is surprising that this complex is air stable.

10 The complexes of the present invention can be used to form electroluminescent devices.

The electroluminescent devices of the invention comprise a transparent substrate which is a conductive glass or plastic material which acts as the anode, preferred substrates are conductive glasses such as indium tin oxide coated glass, but any glass  
15 which is conductive or has a conductive layer can be used. Conductive polymers and conductive polymer coated glass or plastics materials can also be used as the substrate. The electroluminescent material can be deposited on the substrate directly by evaporation from a solution of the material in an organic solvent. The solvent which is used will depend on the material for example alcohols such as ethanol,  
20 ketones such as acetone and methyl acetylacetonate, and chlorinated hydrocarbons such as dichloromethane are suitable in many cases.

Alternatively the material can be deposited by spin coating or by vacuum deposition from the solid state e.g. by sputtering or any other conventional method can be used.

25

In one embodiment of the invention there is a hole transporting layer deposited on the transparent substrate and the electroluminescent material is deposited on the hole transporting layer. The hole transporting layer serves to transport; holes and to block the electrons, thus preventing electrons from moving into the electrode without

- 5 -

recombining with holes. The recombination of carriers therefore mainly takes place in the emitter layer.

5      Hole transporting layers are used in polymer electroluminescent devices and any of the known hole transporting materials in film form can be used.

The hole transporting layer can be made of a film of an aromatic amine complex such as poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl)-1,1'-biphenyl -4,4'-diamine (TPD), polyaniline etc.

10

Optionally dyes such as fluorescent laser dyes, luminescent laser dyes etc. can be included to modify the colour spectrum of the emitted light.

15

Preferably the electroluminescent material is mixed with an inert polymeric material such as a polyolefin e.g. polyethylene, polypropylene etc. and preferably polystyrene. Preferred amounts of the electroluminescent material in the mixture is from 95% to 5% by weight of active material and more preferably 25 to 20% by weight.

20

The hole transporting compound can optionally be mixed with the electroluminescent material in a ratio of 5-95% of the electroluminescent material to 95 to 5% of the hole transporting compound. In another embodiment of the invention there is a layer of an electron injecting material between the cathode and the electroluminescent material layer, this electron injecting material is preferably a metal complex such as a metal quinolate e.g. an aluminium quinolate which will transport electrons when an electric current is passed through it. Alternatively the electron injecting material can be mixed with the electroluminescent material and co-deposited with it.

25

30      In a preferred structure there is a substrate formed of a transparent conductive material which is the anode on which is successively deposited a hole transportation

- 6 -

layer, the electroluminescent material layer and an electron injection layer which is connected to the anode. The anode can be any low work function metal e.g. aluminium, calcium, lithium, silver/magnesium alloys etc.,

5

The preparation of compounds of the invention are shown in the examples

#### Example 1

10

Mono(bathophenanthroline)thorium(IV)chloride. Thorium(IV)chloride (5 mmol, 1.87 g) was dissolved in ethanol/water mixture (2:1 v/v) (75 ml) at 50°C. Bathophenanthroline (5 mmol, 1.66 g) was dissolved in a mixture of ethanol/dichloromethane (2:1 v/v) (75 ml) and added portionwise to the solution of the thorium salt. The mixture was reduced on a hotplate at 100°C over one hour. The precipitate was filtered to give an off-white solid which was washed with diethylether (2 x 25 ml) and dried in vacuo to yield the product (1.9 g).

15

#### Example 2

20

Mono( $\alpha'$ ,  $\alpha''$ ,  $\alpha'''$  tripyridyl)yttrium(III) chloride. Yttrium(III) chloride (2 mmol, 0.61 g) was dissolved in ethanol (100 ml) and  $\alpha'$ ,  $\alpha''$ ,  $\alpha'''$  tripyridyl (2 mmol, 0.47 g) was added. The reaction mixture was warmed for 60 minutes at 50°C and the solvents removed. The residue was washed with diethylether (2x25 ml) and dried in vacuo to give the product (0.80g).

25

#### Example 3

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Tris(2,2,6,6-tetramethyl-3,5-heptanedionato)yttrium(III) mono( $\alpha'$ , $\alpha''$ , $\alpha'''$  tripyridyl). The tris-chelate (1 mmol, 0.64 g) was dissolved in ethanol (100 ml) and  $\alpha'$ ,  $\alpha''$ ,  $\alpha'''$  tripyridyl (1 mmol, 0.23g) was added. The reaction mixture was warmed for 60 minutes at 50°C and the solvents removed. The residue was washed with  
5 diethylether (2x25 ml) and dried in vacuo to give the product (0.50 g). Yield 57%.

#### Example 4

Mono( $\alpha'$ ,  $\alpha''$ ,  $\alpha'''$  tripyridyl)gadolinium(III) chloride. Gadolinium(III) chloride  
10 (0.37 g, 1 mmol) was dissolved in ethanol (150 ml) and  $\alpha'$ ,  $\alpha''$ ,  $\alpha'''$ -tripyridyl (0.23 g, 1 mmol) was added. The reaction mixture was heated under reflux for 1 hour and the solvent removed in vacuo to give the gadolinium adduct (Yield 0.50 g).

#### Example 5

15

Bis(2,2,6,6-tetramethyl-3,5-heptanedionato)europium(II). The reaction was carried out under anhydrous conditions using dried glassware under dry nitrogen and using acetyl nitrile freshly distilled over phosphorus pentoxide. Europium(II) chloride (1.0 g, 5 mmol) was placed in a 250 ml three-neck round-bottom flask fitted with a  
20 condenser, two dropping funnels and nitrogen bubbler. Deoxygenated solution of the diketone (1.84 gms 10 mmol) in acetyl nitrile (50 ml) was placed in the first dropping funnel and deoxygenated acetyl nitrile (150 ml) was placed in the second dropping funnel. Both funnels were under nitrogen and fitted with nitrogen balloons. Acetyl nitrile was allowed to run into the flask and the mixture stirred at 50°C (oil bath) until  
25 dissolution. The diketone solution was then added to the flask and the reaction mixture refluxed for one hour and allowed to cool under nitrogen overnight. The dry diethylether and dried in vacuo at 60°C. (Yield 0.82 g). The filtrate was dried to give pale yellow tris(2,2,6,6-tetramethyl-3,5-heptaneionato)europium(II). (Yield 0.90 g).

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Electroluminescent devices were fabricated and tested.

#### I. Device Fabrication

5

An ITO coated glass piece ( $1 \times 1 \text{ cm}^2$  cut from large sheets purchased from Balzers, Switzerland) had a portion etched out with concentrated hydrochloric acid to remove the ITO and was cleaned and placed on a spin coater (CPS 10 BM, Semitec, Germany) and spun at 2000 rpm for 30 seconds, during which time the solution of the electroluminescent compound was dropped onto it dropwise by a pipette.

10

Alternatively the electroluminescent compound was vacuum evaporated onto the ITO coated glass piece by placing the substrate in a vacuum coater and evaporating the electroluminescent compound at  $10^{-5}$  to  $10^{-6}$  torr onto the substrate.

15

The organic coating on the portion which had been etched with, the concentrated hydrochloric acid was wiped with a cotton bud.

20

The coated electrodes were stored in a vacuum desiccator over calcium sulphate until they were loaded into a vacuum coater (Edwards,  $10^{-6}$  torr) and aluminium top contacts made. The active area of the LED's was  $0.08 \text{ cm}^2$  by  $0.1 \text{ cm}^2$  the devices were then kept in a vacuum desiccator until the electroluminescence studies were performed.

25

The ITO electrode was always connected to the positive terminal. The current vs. voltage studies were carried out on a computer controlled Keithly 2400 source meter.

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Electroluminescence spectra were recorded by means of a computer controlled charge coupled device on Insta Spec photodiode array system model 77II2 (Oriel Co., Surrey, England)

## 5 2. Photoluminescence Measurements

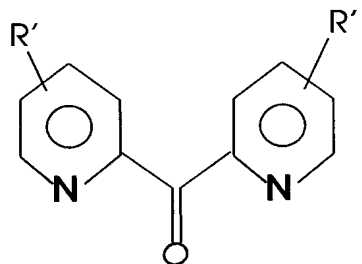
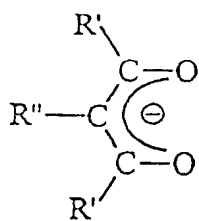
Photoluminescence was excited using 325nm line of Liconix 4207 NB, He/Cd laser. The laser power incident at the sample ( $0.3\text{mWcm}^{-2}$ ) was measured by a Liconix 55PM laser power meter. The radiance calibration was carried out using Bentham radiance standard (Bentham SRS8, Lamp current 4,000A, calibrated by National Physical laboratories, England. The PL studies were carried out on samples or films. The Complexes of the examples were tested and the results shown in the Table and the Spectra attached as Figs. 2 to 6

15 Table

Example	PL %	$\lambda_{\text{max}}/\text{nm}$	CIE		Colour
			x	y	
1	1.0	450	0.17	0.15	Purple
2	6.0	410,520	0.21	0.32	Greenish Blue
3	0.03	460	0.21	0.29	White
4	16	320,450			Purple
5	0.9	420	0.18	0.05	Purple

## Claims

- 5 1. A photoluminescent compound which comprises an organic complex of a transition metal, a lanthanide or an actinide and an organic ligand which photoluminescent compound emits light in the blue or purplish blue spectrum.
- 10 2. An electroluminescent compound which comprises an organic complex of a lanthanide or an actinide and an organic ligand which electroluminescent compound emits light in the blue or purplish blue spectrum when an electric current is passed through it.
- 15 3. A compound as claimed in claim 1 or 2 which comprises a complex of thorium (IV), yttrium (III), gadolinium (III), europium (II), terbium(IV), cerium(IV) and cerium (III) or a mixture of one or more of these.
4. A compound as claimed in claim 1, 2 or 3 in which the ligand is selected from



or

20

- where R' maybe the same or different at different parts of the molecule and each of R'' and R' is a substituted or unsubstituted aromatic or heterocyclic ring structure or a hydrocarbyl or a fluorocarbon or R'' is fluorine or hydrogen or R'' is copolymerised
- 25 with a monomer or is an alkyl group preferably a -C(CH<sub>3</sub>) group, or is selected from

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TMHD,  $\alpha$ ,  $\alpha'$ ,  $\alpha''$ ,  $\alpha'''$  tripyridyl, bathophen (4,7-diphenyl-1,10-phenanthroline), crown ethers and cryptans.

5 5. A compound as claimed in claim 4 in which the ligand is selected from thorium (IV) bathophen, yttrium (III) tripyridyl and yttrium (III) TMHD, and europium (II) TMHD complexes.

6.  $\text{Eu(II)(TMHD)}_2$ .

10 7. A composition which comprises an inert polymer and from 5% to 95% by weight of an electroluminescent compound as claimed in any one of the preceding claims.

15 8. An electroluminescent device which comprises a transparent substrate on which is deposited an electroluminescent compound as claimed in any one of the preceding claims.

9. An electroluminescent device as claimed in claim 8 in which the transparent substrate comprises a conductive glass or plastic material which acts as the anode.

20 10. An electroluminescent device as claimed in claim 9 in which the transparent substrate comprises an indium tin oxide coated glass.

25 11. An electroluminescent device as claimed in any one of claims 8 to 10 in which there is a hole transporting layer deposited on the transparent substrate and the electroluminescent material is deposited on the hole transporting layer.

30 12. An electroluminescent device as claimed in claim 11 in which there is a hole transporting material mixed with the electroluminescent material in a ratio of 5 to 95% of the electroluminescent material to 95 to 5% of the hole transporting compound.

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13. An electroluminescent device as claimed in claim 12 in which the hole transporting layer is an aromatic amine complex.

5 14. An electroluminescent device as claimed in claim 13 in which the hole transporting layer is poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl) -1,1' -biphenyl -4,4'-diamine (TPD) or polyaniline.

10 15. An electroluminescent device as claimed in any one of claims 8 to 14 in which there is a metal anode in contact with the electroluminescent material.

15 16. An electroluminescent device as claimed in any one of claims 8 to 15 in which there is a layer of an electron injecting material between the cathode and the electroluminescent material layer

17. An electroluminescent device as claimed in any one of claims 8 to 16 in which an electron injecting material is mixed with the electroluminescent material and co-deposited with it.

20 18. An electroluminescent device as claimed in claim 16 or 17 in which the electron injecting material is a metal complex or oxadiazole or an oxadiazole derivative.

25 19. An electroluminescent device as claimed in claim 18 in which the electron injecting material is an aluminium quinolate or 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4 oxadiazole.

20. An electroluminescent device as claimed in any one of claims 8 to 19 in which there is a dye incorporated in the electroluminescent layer.

30 21. An electroluminescent device as claimed in 20 in which the dye is a fluorescent

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laser dye or an electroluminescent laser dye.

22. An electroluminescent device as claimed in any one of the preceding claims 8 to 20 in which the anode is a metal.

5

23. An electroluminescent device as claimed in claim 22 in which the anode is a aluminium, magnesium, lithium, calcium or a magnesium silver alloy.

10

24. An electroluminescent device as claimed in any one of the preceding claims in which there are a plurality of layers of electroluminescent material.

25. An electroluminescent device as claimed in any one of the preceding claims in which the layer of electroluminescent material is formed of two or more different electroluminescent compounds.

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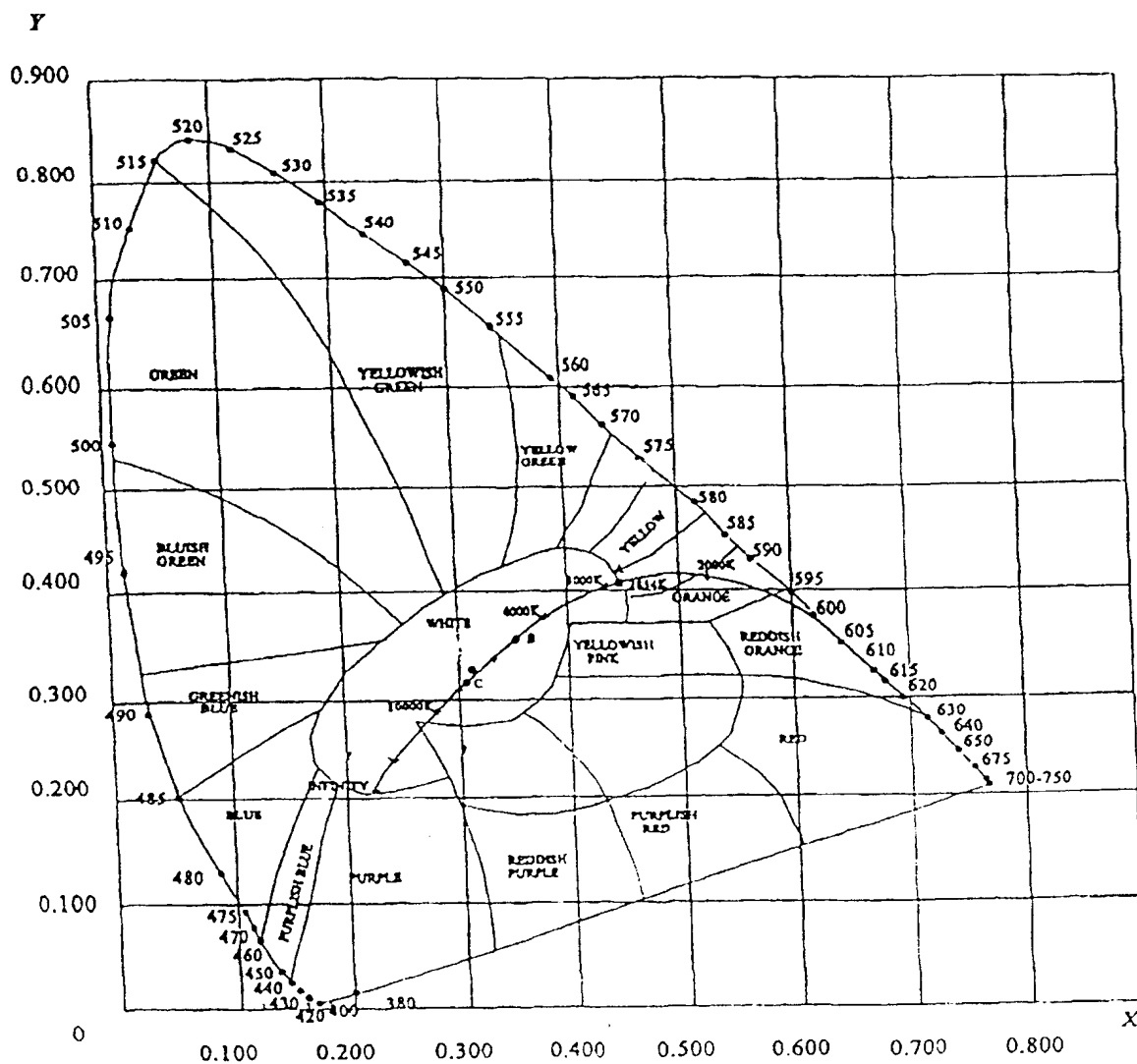


FIG. 1

CIE 1931 x,y chromaticity diagram showing approximate position of perceived colours

2/6

## Example 1

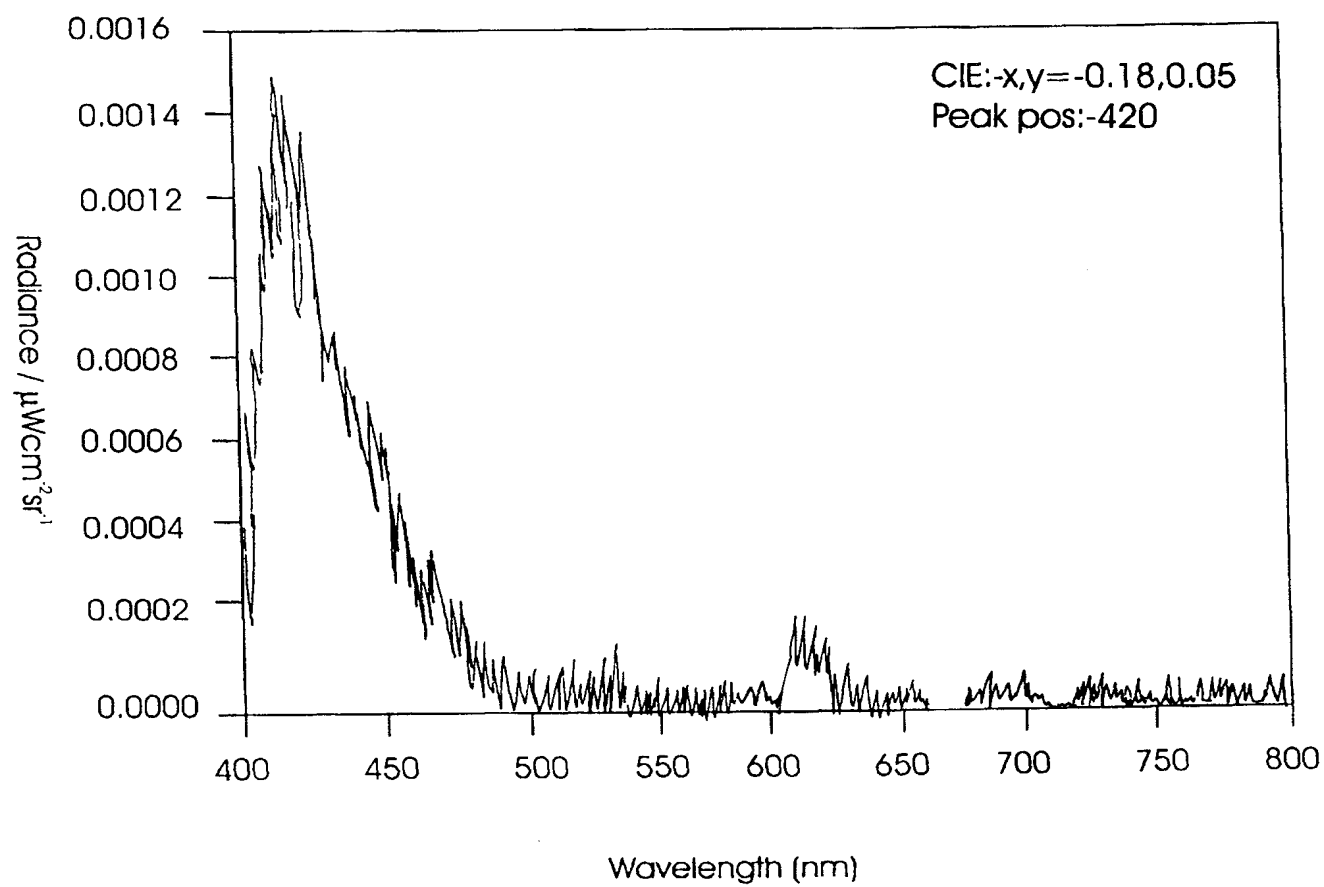


Fig. 2

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## Example 2

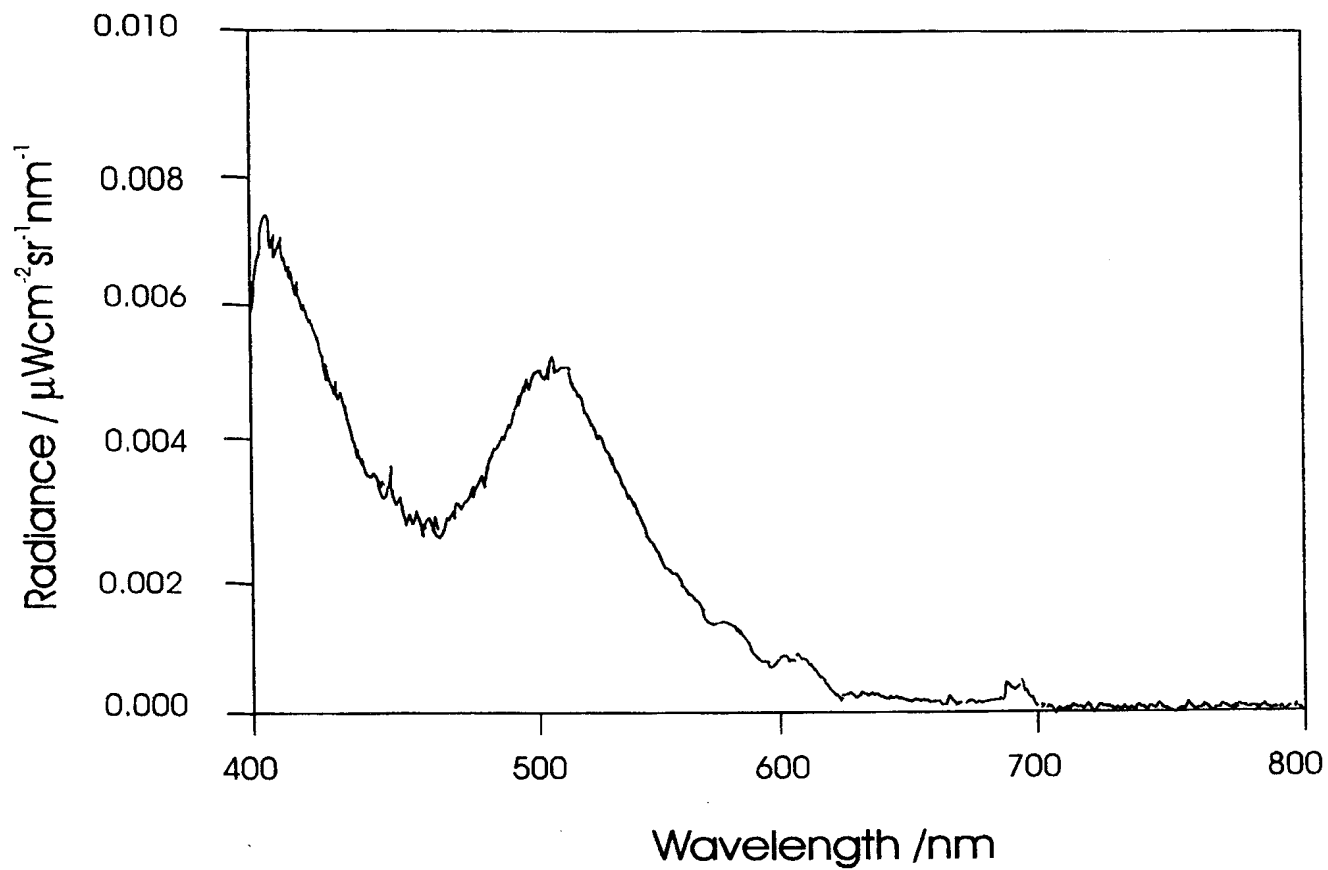


Fig. 3

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## Example 3

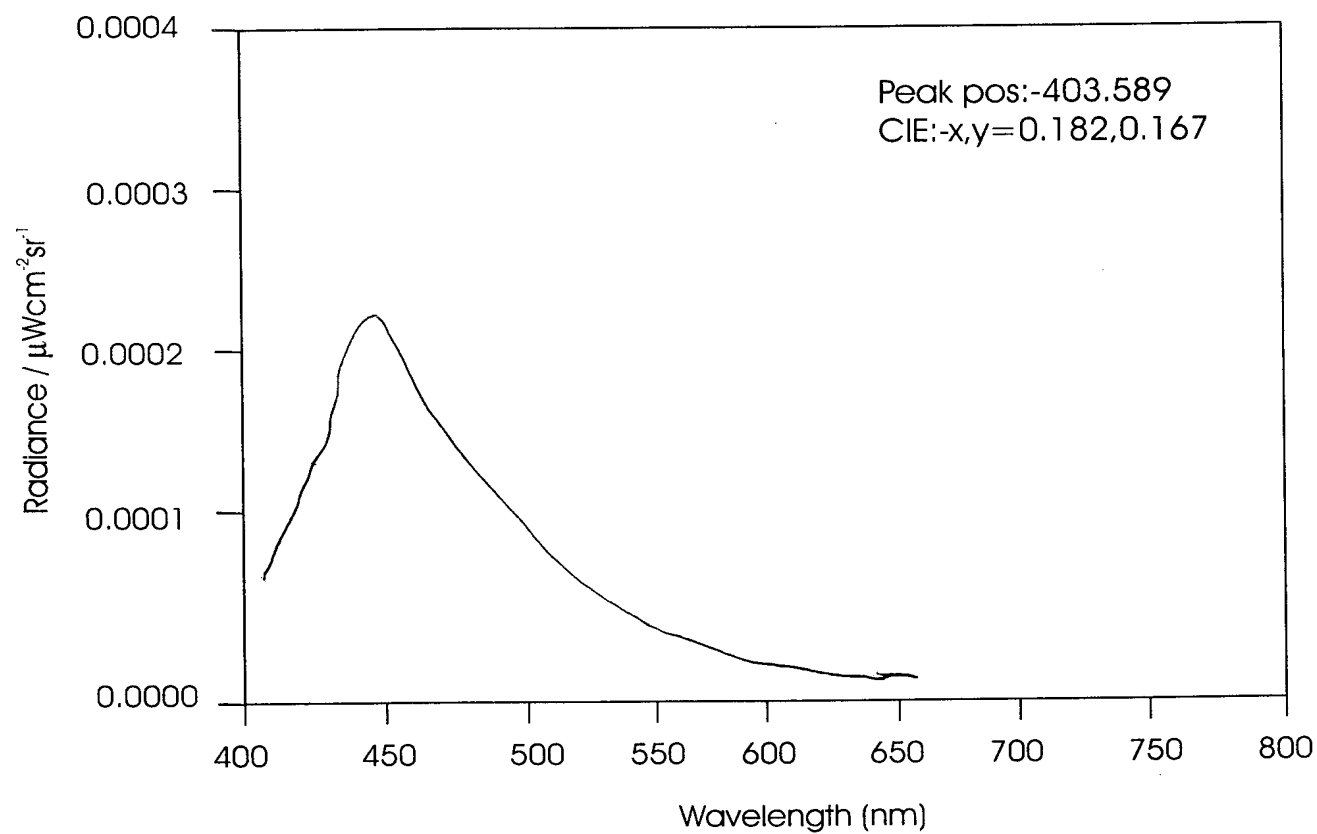


Fig. 4

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## Example 4

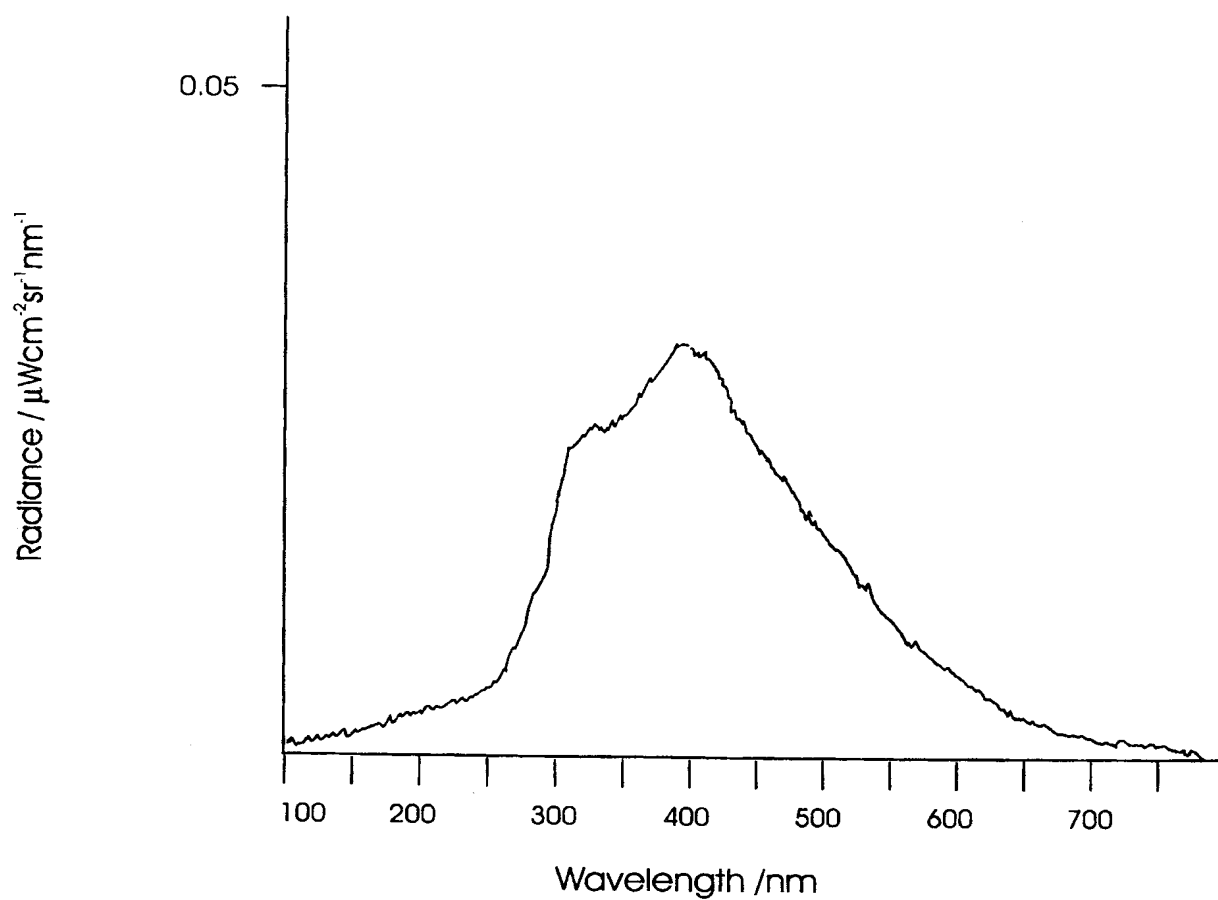


Fig. 5

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## Example 5

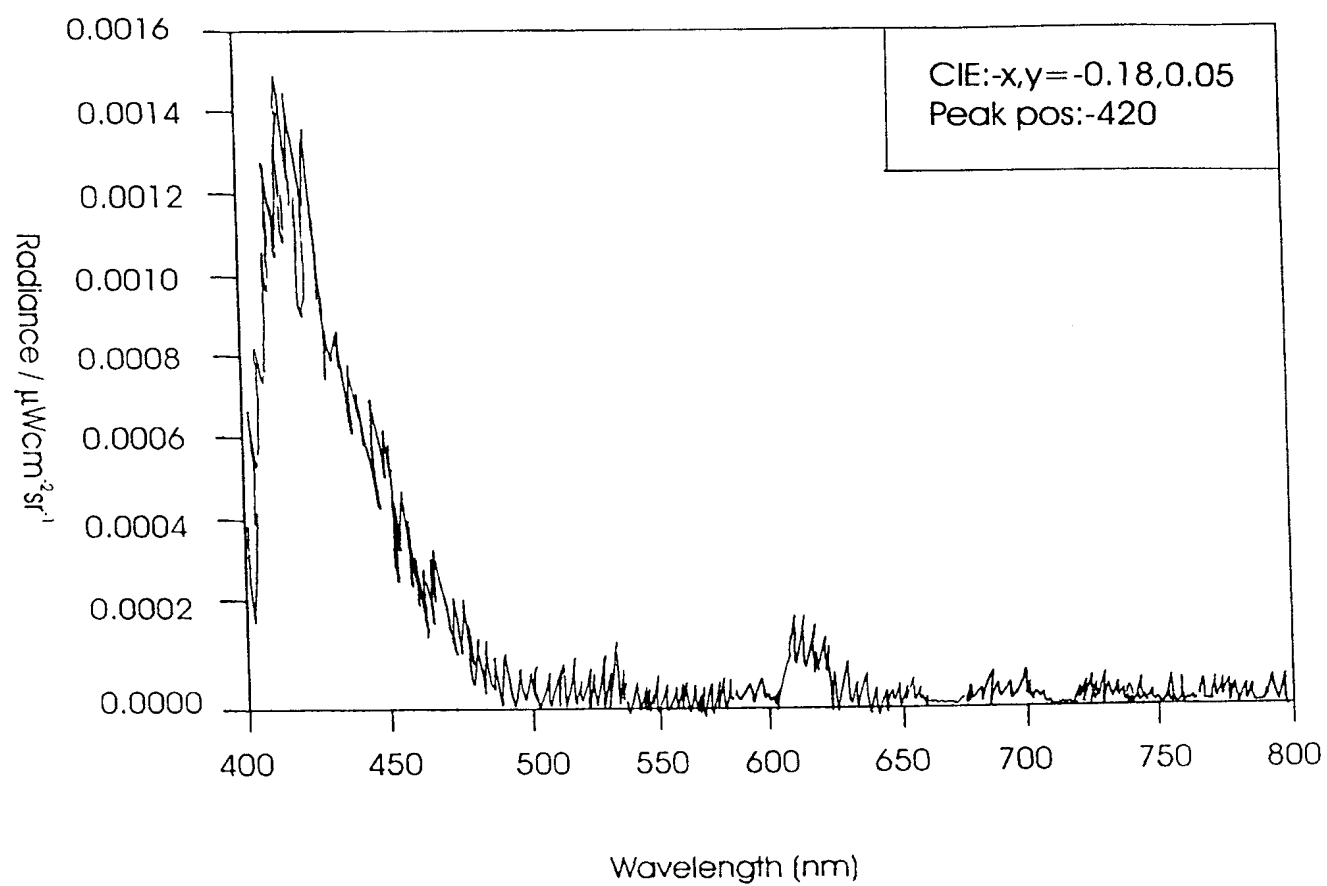


Fig. 6

# INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB 99/03619

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C09K11/06 H05B33/14 H01L51/20

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

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Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	KIDO, J. ET AL: "Organic electroluminescent devices using lanthanide complexes. II" KIDORUI (1995), 26, 110-11, XP002132346 abstract; figure 1 ---	1,2,4, 7-25
X	HAMADA Y ET AL: "BLUE ELECTROLUMINESCENCE IN THIN FILMS OF AZOMETHIN-ZINC COMPLEXES" JAPANESE JOURNAL OF APPLIED PHYSICS, JP, PUBLICATION OFFICE JAPANESE JOURNAL OF APPLIED PHYSICS. TOKYO, vol. 32, no. 4A, PART 02, 1 April 1993 (1993-04-01), pages L511-L513, XP000414014 ISSN: 0021-4922 page L512, column 1, paragraph 2; table 1 --- -/--	1,8-24

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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Shade, M

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International Application No

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## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	KEPERT, CAMERON J. ET AL: "Structural systematics of rare earth complexes. V. The hydrated 1:1 adducts of 2,2':6',2''-terpyridine with the lanthanoid(III) chlorides" AUST. J. CHEM. (1994), 47(2), 365-84 , XP002132347 page 367-368; structures (vi) and (xv) ----	1-5
X	US 5 757 026 A (BURROWS PAUL EDWARD ET AL) 26 May 1998 (1998-05-26) column 9, line 3 - line 5 ----	1,2
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